7. ANALYTICAL METHODS

The purpose of this chapter is to describe the analytical methods that are available for detecting, measuring, and/or monitoring americium, its metabolites, and other biomarkers of exposure and effect to americium. The intent is not to provide an exhaustive list of analytical methods. Rather, the intention is to identify well-established methods that are used as the standard methods of analysis. Many of the analytical methods used for environmental samples are the methods approved by federal agencies and organizations such as EPA and the National Institute for Occupational Safety and Health (NIOSH). Other methods presented in this chapter are those that are approved by groups such as the Association of Official Analytical Chemists (AOAC) and the American Public Health Association (APHA). Additionally, analytical methods are included that modify previously used methods to obtain lower detection limits and/or to improve accuracy and precision.

7.1 BIOLOGICAL MATERIALS

Entry of americium into the human body can be gained through ingestion, inhalation, or penetration through skin. The quantities of americium within the body can be assessed through the use of bioassays that are comprised of *in vivo* measurements and/or *in vitro* measurements. *In vivo* measurements can be obtained through techniques that directly quantify internally deposited americium (using, e.g., a whole body counter). Conversely, *in vitro* measurements provide an estimate of internally deposited americium, utilizing techniques that measure americium in body fluids, feces, or tissue. Examples of these analytical techniques are given in NCRP Report No. 87 (1987) and are also listed in Table 7-1.

7.1.1 Internal Americium Measurements

In vivo measurement techniques are the most direct and widely used approach for assessing the burden of many radioisotopes, including americium, within the body. The *in vivo* measurement of americium within the body is performed with various radiation detectors and associated electronic devices that are collectively known as whole body counters. These radiation detectors commonly utilize sodium iodide (NaI), phoswich (NaI and CsI sandwich), and hyperpure germanium to detect the 59.5 keV gamma-ray that ^{241}Am promptly emits in 35.9% of its alpha decays to ^{237}Np (DOE 1979b; Palmer et al. 1983). Because the attenuation half thickness for the 59.5 keV gamma-ray is 3.5 cm for soft tissues and 1.3 cm for bone, ^{241}Am that has been deposited into specific organs or tissues, such as the lungs, liver, bones, or lymph nodes, can be detected and quantified using whole body counting techniques that appropriately

Table 7-1. Analytical Methods for Determining Americium in Biological Samples

Sample matrix	Sample preparation	Analytical method	Detection limit	Accuracy	Reference
Urine	None	Gamma-ray spectrometry (phoswich detector)	0.08 nCi/200 cm^3	No data	Ide et al. 1985
Urine	Co-precipitation with oxalate	α-Spectrometry	0.08 pCi/80 cm^3	No data	Ide et al. 1985
Urine	None	Gamma-ray spectrometry	0.04 pCi/cm^3	95% at 0.04 pCi/cm^3	Guilmette 1986
Urine	Sample wet ashed, treated with HNO_3 and H_2O_2	α-Liquid scintillation	0.7 pCi/125 cm^3	95% at 0.01–1,000 nCi	Guilmette and Bay 1981
Urine	Sample wet ashed, purified by solvent extraction	Liquid scintillation	14 pCi/100 cm^3	96% at 20,000 dpm spike	Ham et al. 1977
Urine	Sample with ^{241}Am spike co-precipitated with $CaHPO_4$ then with oxalate, purification by diglycol succinate column	α-Liquid scintillation	0.02 pCi/L	96% at 20 dpm spike	Hafez and Hafez 1992
Urine	Sample cleaned-up by co-precipitation, treated with HNO_3 and H_2O_2, wet ashed	Biphasic liquid scintillation	1 pCi/200 cm^3	84%	Bomben et al. 1994
Urine	Spiked sample clean-up by co-precipitation, purified by TRU-spec column and electrodeposition	α-Spectrometry	0.016 pCi/800 cm^3	95% at 0.1–100 pCi/sample	Goldstein et al. 1997

Table 7-1. Analytical Methods for Determining Americium in Biological Samples (*continued*)

Sample matrix	Sample preparation	Analytical method	Detection limit	Accuracy	Reference
Soft tissue	Sample wet ashed, spiked with ^{243}Am, purified by anion exchange, solvent extraction, and electrodeposition	α-Spectrometry	No data	98%	McInroy et al. 1985
Soft tissue	Spiked sample wet ashed, treated with HNO_3/H_2O_2, purified by A-CU column, anion exchange, TRU-spec column, and electrodeposition	α-Spectrometry	No data	53%	Qu et al. 1998
Soft tissue	Sample wet ashed, purified by solvent extraction	Liquid scintillation	1.3 pCi/100 cm^3	96% at 20,000 dpm spike	Ham et al. 1977
Soft tissue	Sample wet ashed, purified by solvent extraction	α-Liquid scintillation	0.7 pCi/g	99% at 3,000 dpm spike	Guilmette and Bay 1981
Bone	Sample wet ashed, purified by solvent extraction	α-Liquid scintillation	0.7 pCi/g	99% at 3,000 dpm spike	Guilmette and Bay 1981
Bone	Sample wet ashed, spiked with ^{243}Am, and purified by anion exchange resin column, solvent extraction, and electrodeposition	α-Spectrometry	No data	98%	McInroy et al. 1985
Feces	Sample wet ashed, purified by solvent extraction	Liquid scintillation	13 pCi/g	91% at 20,000 dpm spike	Ham et al. 1977

Table 7-1. Analytical Methods for Determining Americium in Biological Samples (*continued*)

Sample matrix	Sample preparation	Analytical method	Detection limit	Accuracy	Reference
Feces	Sample wet ashed, purified by solvent extraction	α-Liquid scintillation	1.3 pCi/sample	96% at 3,000 dpm spike	Guilmette and Bay 1981
Feces	None	Phoswich detector	0.02–0.09 nCi/200g	No data	Kramer et al. 1989
Teeth	Sample dissolved in HNO_3, purified by TRU-spec column and electrodeposition	α-Spectrometry	2.7 fCi/sample	98%	Culot et al. 1997
Whole organs and tissues	Animal placed, backbone down in lucite box and positioned 33 cm from NaI(Tl) crystal. Livers and other tissues were counted between two NaI(Tl) crystals.	Gamma-ray spectrometry	No data	No data	Lloyd et al. 1970

TRU = transuranic

account for internal shielding (Graham and Kirkman 1983; Palmer and Rhoads 1989; Palmer et al. 1983). Many configurations of the whole body counter have been utilized, ranging from the more common chest detectors to linear whole body scanners that can be utilized to assess the regional distributions of ^{241}Am over the entire length of the body (Palmer et al. 1983; Toohey and Essling 1980). Where appropriate, shielding of the room that houses the whole body counter and/or the detector is often used to increase the detection sensitivity of the equipment by minimizing background radiation. Also, *in vitro* measurements of americium (see Section 7.1.2) are often used in conjunction with whole body counting when monitoring individuals working with americium.

Calibration of whole body counters is achieved through the use of tissue-equivalent phantoms that are constructed to mimic the shape and density of the anatomical structure (e.g., the human torso), using tissue equivalent materials such as polystyrene or epoxies. In some phantoms, a human rib cage is added to account for the attenuation of gamma-rays by bone in the whole body counts (DOE 1979b). Americium standards are inserted or molded into the phantom at locations where this isotope is expected to accumulate, such as in the lung, liver, or bone. Comparisons of the activity obtained from the phantom to the known activity of the americium standards are used to determine the efficiency of the counting technique and, thus, provide the basis for calibrating the technique. Calibration of whole body counts can be further refined by obtaining actual anatomical measurements of the individual to be scanned. For example, chest wall measurements using ultrasound techniques are used to account for the variability in attenuation that results from individual differences in the chest wall thickness and improving the calibration of chest counts of americium (DOE 1979b). Another approach to refining the calibration of whole body counters is the comparison of external measurements to the actual americium content in organs and bone of cadavers (Palmer et al. 1985). These refinements in calibration phantoms can lead to a more accurate assessment of the total body or organ burden of americium.

The NRC requires that the occupational intake of americium isotopes not exceed certain specified Annual Limits on Intake (ALIs) for the inhalation and oral routes of exposure. For ^{241}Am and ^{243}Am, the oral ingestion ALI is 0.8 μCi and the inhalation ALI is 0.006 μCi, both of which are based on the deterministic dose limit to the bone surface (NRC 2000).

In assessing initial exposure, whole body counting techniques measure amounts of americium that have been deposited within organs or tissues and have not been excreted. In cases of accidental ingestion, some of the americium may have been excreted in the urine or through the feces, before exposure is assessed. In particular, soluble forms of americium are thought to be readily excreted through the urine.

Long-term assessment of americium burden within an individual can be complicated by the mobilization of americium from the original site of deposition (e.g., the lung) to other sites within the body, such as the liver or bone (Fry 1976). This can lead to overestimates of the overall total body burden of americium depending on which regions the americium counts are obtained (Kathren et al. 1988). Additionally, the retention (biological half-life) of americium within the body can vary greatly between individuals (Fry 1976). Also, direct comparisons of americium body burdens and clearance rates between laboratories can be complicated by the differing whole body measurement techniques, calibration methods, and methods used to account for normal background radiation counts that are utilized within the different laboratories (DOE 1979b). These limitations are largely resolved by performing periodic internal monitoring and adjusting the model parameters to account for the individual's actual distributed retention.

7.1.2 External Radiation Measurements

In vitro analyses of americium are routinely performed in situations where *in vivo* analyses cannot be obtained or in support of an *in vivo* monitoring program. Urine is the preferred sample for *in vitro* analyses of americium, although other sample types, such as feces, tissue, bone, or blood, can also be used on a more limited basis. Urine provides for an analysis of soluble or transportable americium, fecal analysis can be used to measure ingestion or clearance of americium, and tissue is used to assess whole or regional body burdens of americium (Guilmette and Bay 1981; Ide 1986; Ide et al. 1985; McInroy et al. 1985).

Urinalysis for americium can be performed on the basis of total either mass or total activity. There are a number of methods that have the selectivity and/or sensitivity to measure americium in biological matrices including spectrophotometry, fluorometry, mass spectrometry (MS), and radioassays (Dacheux 1998; Hafez and Hafez 1992; Poupard and Jouniaux 1990; Thouvenout et al. 1993). Of these methods, radioassays (e.g., gross alpha, analysis alpha spectroscopy, gamma-ray spectroscopy, liquid scintillation techniques) are preferred because of their ease of use, detection sensitivity, and rapidity of analysis (Alvarez and Navarro 1996; Dacheux and Aupiais 1997; Guilmette 1986). These methods typically involve a preliminary concentration step and wet/dry ashing of the sample that is often followed by an oxidation of the radionuclides in the sample residue. To remove the possible interferences of other alpha-emitters, ion exchange, co-precipitation, and adsorption techniques are applied to the purified sample before alpha or liquid scintillation techniques are applied (see Table 7-1). Radioassays can also be applied to the measurement of americium in fecal, tissue, and bone samples using methods that are similar to those described for urinalysis, except for some additional purification and extraction steps that are required to remove interfering materials such as iron or excess mass (Guilmette and Bay 1981; Hafez and Hafez 1992; Ham et al. 1977; Qu et al. 1998).

Of the radioassays that are commonly used to quantify americium, α-spectroscopy is used when isotopic analyses of americium must be conducted (e.g., ^{241}Am and ^{243}Am). ^{243}Am is often added as a tracer to estimate the efficiency of the sample preparation method when quantifying ^{241}Am in biological matrices. The α-spectroscopy technique differentiates between the two americium isotopes based on the difference in the energies of the alpha particles that are emitted from ^{241}Am and ^{243}Am, and then quantifies the amounts present in the sample considering the activity that each contributes, its detection efficiency, and the frequency with which the individual alpha particles are emitted. Mass spectrometric techniques are also capable of isotopic quantification of americium (Dacheux and Aupiais 1998; Poupard and Jouniaux 1990). These techniques are more rapid than the α-spectroscopy detection method, but the cost has been much higher. However, the cost of mass spectrometers has decreased in recent years, making the cost of analyzing samples by mass spectroscopy and α-spectroscopy more comparable. Higher sensitivity can also be achieved with α-spectroscopy by resorting to long sampling times, sometimes referred to as time-averaging.

Accuracy of *in vivo* and *in vitro* measurements of americium is determined through the use of standard, certified radioactive sources with known concentrations of americium. The primary source of certified americium standards is the National Institute of Standards and Technology (NIST). Standard solutions are available for ^{241}Am (SRM 4322, 40 Bq/g [1.1 nCi/g]) and ^{243}Am (SRM 4332, 40 Bq/g [1.1 nCi/g]). Standard Reference Materials for human lung (SRM 4351) and human liver (SRM 4352) are also available from NIST.

7.2 ENVIRONMENTAL SAMPLES

There are two common approaches for measuring americium in the environment. Americium can either be measured directly in the field (*in situ*) using portable survey instruments or samples can be procured from the field and returned to the laboratory for quantification of americium.

7.2.1 Field Measurements of Americium

In situ measurement techniques are extremely useful for the rapid characterization of radionuclide contamination in the environment, such as soils, sediments, and vegetation, or when monitoring personnel for exposure to radionuclides. Information regarding field measurement methods, minimum detectable concentrations, and soil-to-plant concentration factors is available for various radionuclides, including americium (MARSSIM 2000; NRC 1992, 1998b). The measurement of radionuclides in the environment is conducted with portable survey instruments that are equipped with α-scintillators or gamma-ray spectrometers. However, the use of gamma-spectrometers in field survey equipment is preferred for measuring americium in the field because the low-energy gamma-ray photon that is emitted during the decay of ^{241}Am is more penetrating and measurements are less affected by the presence of vegetation and surrounding soil than is the alpha particle that is also emitted (Byrne and Komosa 1993). This provides the advantage for assessing the level of americium both on and below the surface (e.g., up to 3-cm depth in some soils). These gamma-ray spectrometers are equipped with either a thin sodium iodide or a high purity germanium detector that is able to distinguish the 59.5 keV gamma-ray emitted from ^{241}Am from most environmental gamma-rays emitted from other radionuclides (Fong and Alvarez 1997). Another advantage of these spectrometers is the ability to discriminate the 59.5 keV photons of ^{241}Am from the much lower energy photons emitted from the ^{239}Pu and ^{240}Pu that are usually associated with americium. Minimum detectable activities (MDAs) of 0.4 Bq/g (10 pCi/g) for ^{241}Am are routinely achieved with MDAs as low as 0.04 Bq/g (1 pCi/g) obtained with longer counting times (-30 days).

One of the limitations of the portable field survey instruments in the measurement of americium is that their quantitative accuracy depends on how well the lateral and vertical distribution of americium in the soil compares with the calibration parameters used. These methods can provide a rapid assessment of americium levels on or below surfaces in a particular environment; however, laboratory-based analyses of samples procured from these environmental surfaces must be performed in order to ensure accurate quantification of americium (and other radionuclides). This is due, in part, to the strong self absorption of the 59.5 keV gamma-ray by environmental media, such as soil. Consequently, the uncertainty in the depth distribution of americium and the density of the environmental media may contribute to a >30% error in the field survey measurements. Currently, refinements in calibration strategies are being developed to improve both the precision and accuracy (10%) of gamma-ray spectroscopy measurements of americium within contaminated soils (Fong and Alvarez 1997).

7.2.2 Laboratory Analysis of Environmental Samples

Analytical methods for quantifying americium in environmental samples are summarized in Table 7-2. The methods that are commonly used in the analysis of americium based on activity are gross α analysis, α-spectrometry and gamma-ray spectrometry. MS detection techniques are used to measure the mass of americium in environmental samples. (The mass-activity conversion factor for ^{241}Am is 0.29 μCi/μg or 3.43 μg/μCi [Harvey et al. 1993]).

The analysis of americium in air is based on the quantification of americium within particulates that become trapped on cellulose or glass fiber filters after a calibrated amount of air is pulled through the filters. A common method of analysis for americium on a glass fiber filter is a rather complex procedure involving many solvent extraction and column purification steps, followed by electrodeposition and α-spectroscopy. The extensive purification is required to prevent impurities within the sample from absorbing or reducing the energy of emitted alpha particles, termed self-absorption. Alpha-emitting contaminants must also be removed (e.g., ^{238}Pu) from the samples to prevent these materials from interfering with the α-spectrometry measurements of ^{241}Am and ^{243}Am (ASTM 1997; Lovette et al. 1990). Initially, the filter media is dissolved with HNO_3 and H_2O_2, the residue wet ashed with HNO_3, and then purified using anion exchange chromatography, solvent extraction with 50% bis-(2-ethylhexyl) phosphoric acid (HDEHP) in toluene, another passage of the sample through an anion exchange column, and then oxidation of the sample with HNO_3 and H_2O_2. Because ^{243}Am is commonly used as an internal standard when quantifying ^{241}Am, α-spectrometry is utilized to resolve and quantify the americium isotopes in the purified filter sample. Preparation of the purified filter sample for α-spectrometry requires electrodeposition of the americium from a sulfate solution onto a stainless steel or platinum disc from which alpha counts are obtained during the α-spectrometry analysis (DOE 1997b). The accuracy of this method of analysis for americium can vary between 85 and 102% and the minimum detectable activity (MDA) often ranges between 0.032 and 0.023 pCi/sample (Goldstein et al. 1997).

For the analysis of americium in water, there is a broad array of sample preparation and detection methodologies that are available (see Table 7-2). Many of the common and standardized analytical methodologies typically include the minimization of sample volume, purification through co-precipitation, anion exchange column chromatography, and solvent extraction techniques followed by radiochemical detection of americium in the purified sample. Gross alpha analysis or liquid scintillation are common detection techniques that are utilized to quantify americium in these methods. However, if lower detection sensitivity or isotopic determination is required, then α-spectrometry is the preferred

Table 7-2. Analytical Methods for Determining Americium in Environmental Samples

Sample matrix	Sample preparation	Analytical method	Detection limit	Accuracy	Reference
Air	Sample collection on cellulose filter, dry ashed, solvent extracted	Biphasic liquid scintillation	1 pCi	95%	Bomben et al. 1994
Air	Filter wet ashed in HNO_3/HF, purified with cation and anion exchange columns and electrodeposition	α-Spectroscopy	No data	No data	Knab 1979
Air	Cellulose filter dry ashed, dissolved in HNO_3/HF, H_2O_2/$HClO_4$, purified with anion exchange, TRU-spec columns followed by electrodeposition.	α-Spectroscopy	0.023 pCi/sample	102%	Goldstein et al. 1997
Water	Sample fusion with pyrosulfate, precipitated with barium sulfate	Scintillation counter	No data	99.5%	Sill and Williams 1969
Water	Wet ashed, purified by solvent extraction	Biphasic liquid scintillation	1 pCi/sample	95%	Bomben et al. 1994
Water	Treated with HNO_3/H_2O_2, HF/HCl, anion exchange, TRU-spec column, electrodeposition	α-Spectroscopy	0.026 pCi/L	101%	Goldstein et al. 1997
Water	Solvent extracted	PERALS	0.007 pCi/L	104%	Dacheux and Aupiais 1998

Table 7-2. Analytical Methods for Determining Americium in Environmental Samples (*continued*)

Sample matrix	Sample preparation	Analytical method	Detection limit	Accuracy	Reference
Sea water	Co-precipitation with iron hydroxide, purified by anion exchange, co-precipitation with $BiPO_4$, cation exchange, electrodeposition	α-Spectroscopy	No data	64–79%	Lovette et al. 1990
Sediments	Sample fusion with KF and pyrosulfate, coprecipitate with $BaSO_4$	Scintillation counter	No data	No data	Sill and Williams 1969
Sediments	Sample leached with HNO_3/HF, filtered, purified by KL-HDEHP resin columns, solvent extracted, and electrodeposition	α-Spectroscopy	No data	95–99%	Guogang et al. 1998
Sediments	None	Gamma-ray spectroscopy	0.02–0.06 pCi/g	108–118%	Joshi 1989
Soil	Sample fusion with KF and pyrosulfate, coprecipitate with $BaSO_4$	Scintillation counter	No data	No data	Sill and Williams 1969
Soil	Wet ash in HNO_3/HF, purified with cation and anion exchange columns, electrodeposition	α-Spectroscopy	No data	No data	Knab 1979

Table 7-2. Analytical Methods for Determining Americium in Environmental Samples (*continued*)

Sample matrix	Sample preparation	Analytical method	Detection limit	Accuracy	Reference
Soil	Dry ash, digest in HNO_3/HCl, anion exchange, Ca-oxalate and Fe $(OH)_2$ coprecipitation, anion exchange, electrodeposition	α-Spectroscopy	27 pCi/g	75–92%	Sanchez and Singleton 1996
Soil	Sample leached with HNO_3/HF, filtered, purified by KL-HDEHP resin columns, solvent extracted, and electrodeposition	α-Spectroscopy	No data	95–99%	Guogang et al. 1998
Soil	None	Gamma-ray spectroscopy	0.02–0.06 pCi/g	108–118%	Joshi 1989
Vegetation (grasses)	Ashed, HNO_3/HF, precipitation with oxalate and La, anion exchange, solvent extraction	α-Spectroscopy	0.011 pCi/g	No data	Bunzl and Kracke 1990
Vegetation	Ashed, digested with HNO_3-H_2O_2, oxalate and Fe precipitations, anion exchange, solvent extraction, electrodeposition	α-Spectroscopy	0.3 fCi/g	73–109%	Cooper et al. 1993
Vegetation	Ashed, digested with HNO_3-HCl, anion exchange, Ca-oxalate and Fe precipitations, anion exchange, electrodeposition	α-Spectroscopy	27 pCi/g	75–92%	Sanchez and Singleton 1996

Table 7-2. Analytical Methods for Determining Americium in Environmental Samples (*continued*)

Sample matrix	Sample preparation	Analytical method	Detection limit	Accuracy	Reference
Lichen, moss	Ashed, leached with HCl, Microthene-TNOA and KL-HDEHP column extractions, solvent extraction, electrodeposition	α-Spectroscopy	0.9 fCi/g	No data	Jia et al. 1997
Biota	Ashed, digested with HNO_3-H_2O_2, oxalate, and Fe precipitations, anion exchange, solvent extraction, electrodeposition	α-Spectroscopy	0.3 fCi/g	98–100% 480% (shrimp)	Cooper et al. 1993

KL-HDEHP = 50% di(2-ethylhexyl) phosphoric acid, 60-100 mesh resin; PERALS = Photon/electron rejecting alpha liquid scintillation; TNOA = tri-*n*-octylamine; TRU = transuranic method to quantify ^{241}Am, especially when ^{243}Am is used as an internal standard in the radiochemical assay (Dacheux and Aupiais 1997; DOE 1997b; Goldstein et al. 1997; Harvey et al. 1993; Sill and Williams 1969). These detection methods can provide measurements of total activity of americium within a sample, especially when appropriate steps have been taken to purify the sample of interfering materials or minimize the influence of other radionuclides on radiochemical activity (Dazhu et al. 1991). The presence of alpha, beta, and/or gamma emitting radionuclides in the sample can contribute to the counts measured in radiochemical detection methods and, thus, affect the accuracy of the assay for determining the quantity of americium within a sample.

There are methods available to quantify the total mass of americium in environmental samples. Mass spectrometric methods provide total mass measurements of americium isotopes (Dacheux and Aupiais 1997, 1998; Halverson 1984; Harvey et al. 1993) however, these detection methods have not gained the same popularity as is found for the radiochemical detection methods. This may relate to the higher purchase price of a MS system, the increased knowledge required to operate the equipment, and the selection by EPA of α-spectrometry for use in its standard analytical methods. Fluorimetric methods, which are commonly used to determine the total mass of uranium and curium in environmental samples, have limited utility to quantify americium, due to the low quantum yield of fluorescence for americium (Thouvenout et al. 1993).

Several rapid radiochemical analysis techniques have been developed that require either no or minimal sample preparation that can, under optimized conditions, provide the required selectivity and sensitivity to quantify americium in environmental samples, such as soils and sediments (Byrne and Komosa 1993; Cutshall et al. 1983; Guilmette 1986; Joshi 1989). These techniques utilize a phoswich-based low energy photon detector that detects the gamma-rays emitted from ^{241}Am in a neat or ashed environmental or biological sample. On average, these methods have a minimum significant measurable activity (MSMA) of approximately 1 pCi/sample. To achieve this low MSMA, the counting efficiency of the detector must be standardized against the sample size and composition in order to assure the desired accuracy of the assay (Cutshall et al. 1983; Joshi 1989). However, one limitation of the assay is the need to know the isotopic composition of the sample since the x-rays that accompany the decay of other radionuclides may also be counted by the phoswich detector.

Several methods have been described in the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM 2000) for the survey and investigation of sites contaminated with radioactive materials. At the high end of the survey instruments, costing over $1,000,000 in 1995, is an inductively coupled plasma mass spectrometer (ICP-MS) with a laser added to the front end to vaporize small portions of a surface to be analyze (laser ablation), thereby avoiding physical sample collection.

The quantity of americium in soil, sediments, vegetation, and biota is determined using methods similar to those described above. For example, in a standardized method developed by Department of Energy (DOE) (DOE 1997b) soil samples are dissolved with a series of acid treatments (e.g., HNO_3, HF) and initially purified through co-precipitation using calcium oxalate followed by co-precipitation with an Fe carrier. A final purification of the sample is achieved by passing the sample through a series of anion exchange columns (e.g., HNO_3, HCl, and NH_4 SCN), followed by electrodeposition of the americium (chloride form) onto a platinum disc in preparation for α-spectrometry analysis.

In another standardized method developed for the analysis of americium in soil (ASTM 1997), a different approach is taken towards purifying the dissolved soil sample that relies on a series of co-precipitations and solvent extractions to prepare the soil sample for α-spectrometry analysis. After the soil has been dissolved, the sample is initially purified by co-precipitation with barium sulfate followed by solvent extraction of the redissolved precipitate with 15% HDEHP in n-hexane. The extracted trivalent actinides and lanthanides are stripped from the organic phase using nitric acid containing sodium bromate. The subsequent solution containing the trivalent actinides and lanthanides is extracted again with 15% HDEHP in n-hexane to remove plutonium, thorium, and tetravalent curium. The aqueous phase is further purified through co-precipitation with a lanthanum carrier to isolate the rare earth fluorides followed by a treatment using silver nitrate and ammonium persulfate/ammonium fluoride to precipitate all remaining rare earth fluorides, except for hexavalent americium fluoride. The hexavalent americium is reduced back to the trivalent state using hydrogen peroxide and then reprecipitated with a neodymium carrier in preparation for α-spectrometry analysis. Both the DOE and the American Society for Testing and Materials (ASTM) methods of analysis provide good precision (<6% standard derivation) with no statistically significant bias (at the 5% level) observed. Analysis of americium in sediments, vegetation, and biota can also be performed using variations of the abovementioned methods or other methods, as exemplified in Table 7-2.

The detection limits, accuracy, and precision of any analytical methodology, as well as the composition of the sample medium, are important parameters in determining the appropriateness of a method to quantify a specific analyte at the desired level of sensitivity within a particular matrix. The lower limit of detection (LLD) has been adopted to refer to the intrinsic detection capability of a measurement procedure (sampling through data reduction and reporting) to aid in determining which method is best suited for the required sample quantification (NRC 1984). Several factors influence the LLD, including background counting rates, size or concentration of sample, detector sensitivity, recovery of desired analyte during sample isolation and purification, level of interfering contaminants, and, particularly, counting time. Because of these variables, the LLDs between laboratories, utilizing the same or similar measurement procedures, will vary.

The accuracy of a measurement technique in determining the quantity of a particular analyte in environmental samples is greatly dependent on the reliability of the calibrating technique. Thus, the availability of standard, certified radiation sources with known concentrations of americium are required in order to ensure the reliability of the calibration methods and accuracy of americium measurements in environmental samples. The primary source of certified americium standards is the NIST. Standard solutions of ^{241}Am (SRM 4322, 40 Bq/g [1.1 nCi/g]) and ^{243}Am (SRM 4332, 40 Bq/g [1.1 nCi/g]) are available. Standard reference materials are also available from NIST and International Atomic Energy Agency (IAEA) for a number of environmental matrices; for example, soils and sediments (Rocky Flats Soil [SRM 4353], river sediment [SRM 4350B], and Peruvian soil [SRM 4355], sediments [IAEA 367, IAEA 135]).

7.3 ADEQUACY OF THE DATABASE

Section 104(i)(5) of CERCLA, as amended, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of americium is available. Where adequate information is not available, ATSDR, in conjunction with the National Toxicology Program (NTP), is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of americium.

The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP, and EPA. They are defined as substance-specific informational needs that if met would reduce the uncertainties of human health assessment. This definition should not be interpreted to mean that all data needs discussed in this section must be filled. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be proposed.

7.3.1 Identification of Data Needs

Methods for Determining Biomarkers of Exposure and Effect. Analytical methods with satisfactory sensitivity and precision are available to determine the levels of americium in human tissues and body fluids. However, improved methods are needed to assess the biological effects of americium in tissues.

Methods for Determining Parent Compounds and Degradation Products in Environmental Media. Analytical methods with the required sensitivity and accuracy are available for quantification of americium, both total and isotopic, in environmental matrices (see Table 7-2). Knowledge of the levels of americium in various environmental media, along with the appropriate modeling (see Chapters 3 and 5), can be used to evaluate potential human exposures through inhalation and ingestion pathways.

Whether in the environment or in the human body, americium will undergo radioactive decay to form a series of radioactive nuclides that end in a stable isotope of lead (for ^{243}Am) or bismuth (for ^{241}Am) (see Chapter 4). However, more sensitive analytical methods are needed for accurately measuring very low levels of these radionuclides. Practically speaking, since ^{239}Pu (for ^{243}Am) and ^{237}Np (for ^{241}Am) have such extremely long half-lives, 2.41×10^4 and 2.14×10^6 years, respectively, few decay products need to be considered.

7.3.2 Ongoing Studies

In the Federal Research in Progress (FEDRIP) lists an ongoing effort by G. Entine of Radiation Monitoring Devices, Inc., in Watertown, Massachusetts, which proposes development of a new type of larger, high resolution, high efficiency germanium detector in order to provide effective detection, localization, as well as quantification of the radionuclides inhaled by workers. The proposed effort is expected to lead to a simpler, higher performance, and more economical germanium detector system for lung screening. This effort is supported by the National Heart, Lung, and Blood Institute (FEDRIP 2000).

Table 7-3. Ongoing Studies on Analytical Methods for Americium

Investigator	Affiliation	Subject	Sponsor
Entine, G	Radiation Monitoring Devices, Inc., Watertown, Massachusetts	The goal of the proposed effort is to develop a new type of larger, high resolution, high efficiency germanium detector in order to provide effective detection, localization, as well as quantification of the radionuclides inhaled by workers. The proposed effort is expected to lead to a simpler, higher performance, and more economical germanium detector system for lung screening.	National Heart, Lung, and Blood Institute

Source: FEDRIP 2000